14. *On the ALTERATION of COARSELY SPHERULITIC ROCKS.* By
GRENVILLE A. J. COLE, Esq., F.G.S. (Read March 10, 1886.)

[PLATE IX.]

As has been previously stated*, all the phenomena that accompany spherulitic structure on an ordinary scale appear reproduced among lavas that may be called coarsely spherulitic. Ill-defined segregations, differing little from the matrix, the fluidal and banded structures of the rock running indifferently through both, are found measuring an inch or two in diameter, equally with forms of marked radial or concentric character. The scale on which certain processes of decomposition and reconstruction, especially those that affect differently the spherulite and the matrix, are revealed in these bold examples, and the interest attaching to their effect upon the ultimate character of the mass, have led me to bring together a few observations on the alteration of coarsely spherulitic rocks. Some of the finest instances, moreover, may be found in our own British lava-flows of Palæozoic age, and with these the field-worker is not unfrequently called upon to deal.

The well-known black pitchstone of Planitz, near Zwickau in Saxony, which is of Permian age, contains angular aggregates of calcite and chalcedony enclosed in large brown nodules. These remarkable occurrences were referred to by Cotta as early as 1849†, and his figure was copied by Delesse in a comprehensive paper on "Les Roches globuleuses"‡. Cotta considered the nodules surrounding the chalcedony as inclusions of an adjoining porphyry, like those in the pitchstone of Spechthausen; but the microscope proves them to be in reality large brown spherulites, often 3 centims. in diameter, and differing only slightly from the glass. Why the agents of alteration should have attacked them in so marked a degree is indeed a puzzling matter, which will be best solved by a study of the "Lithophysen" of more recent lavas; but there can be little doubt that these spherules were once solid to the core. Since they have no internal radial or concentric structure, the decomposition has hollowed them out irregularly, or has started along and spread from any cracks that might occur. The relation of the shape of the cavity so formed to these preexisting cracks may often be seen at the angular ends of its branches. The hollow has ultimately been filled with chalcedony or with calcite, which may also be found in cracks throughout the rock. There is no sign of a central mineral nucleus from which this material might have been derived, since the lines of flow throughout the nodule do not bend round as when an obstacle is present, but may be connected up with one another straight across the branchings of the secondary mass. The same evidence strongly opposes the supposition of the former existence of

* Q. J. G. S. vol. xli. pp. 165, 166.

† Leitfaden und Vademecum der Geognosie, p. 76.

‡ Mémoires de la Soc. géol. de France, 2me série, tome iv. p. 315.

a central vesicle, produced by the expansion of gases while the lava was still cooling down (Pl. IX. fig. 1).

An experiment as to the degree of solubility in acid of the spherulite and the matrix respectively yielded only an unsatisfactory result. Equal quantities of the powdered substances gave, after eight days' immersion, with two boilings, in strong hydrochloric acid, the following percentages of material dissolved :—

Glassy matrix.	Spherulite.
7.57	8.80

Although the portions of the spherulite were chosen with great care, some of the above difference might be due to minute included veins of calcite. On the other hand it is possible that even such a slight variation in behaviour as that indicated above might be greatly intensified by the temperature, and perhaps pressure, at which the natural solvents worked.

Angular secondary aggregates, apparently very similar to those of Zwickau, occur in other Saxon porphyries and pitchstones, and are recorded, as one might expect, from the most highly altered zones*. These igneous sheets are associated with *Rothliegende* sandstones and conglomerates, which are sometimes cemented by silica, sometimes by carbonate of lime, and which contain occasional calcareous layers; when, therefore, the felsitic spherules had once been hollowed out (a process begun, in all likelihood, while the volcanic centre was still active), the calcite and chalcedony may easily have infiltrated from the surrounding sedimentary rocks.

The process of secondary devitrification† tends often to lessen still further the difference between the spherulites and the matrix; so that we may at any time be confronted by an altered lava containing angular shreds of quartz, chalcedony, &c., which, had we none of the intervening stages, might even be regarded as fragments picked up during flow.

The red rocks of the Wrockwardine (Wrekin) area have preserved their original structures well; but among them is a coarsely spherulitic layer in which the formation of secondary quartz-masses has in places gone to an extreme. As in the historic Zwickau pitchstone and many later examples, the banding often traverses the larger spherules and carries trains of small ones through them. The largest spherulites are sometimes aggregates of those of medium size, and become, indeed, in this way elongate, knotty, or irregular. The development of quartz in their interior has been previously described‡; but I confess that on recently revisiting the area, I was unprepared for the scale on which this alteration has taken place. One spheroidal mass, for example, measures 11 centim. ($4\frac{1}{4}$ inches) in diameter, and yet the residual red material is often not more

* *Lehmann and Siegert. Erläuterungen zur geol. Karte des Königr. Sachsen, Section Hohenstein*, pp. 44, 45, & 53.

† See Bonney, *Proc. Geol. Soc.* 1885, p. 87.

‡ *Q. J. G. S.* vol. xli. p. 167. Cf. Monteiro, "Description minéralogique du Pyroméride globale de Corse." *Journal des Mines*, tome xxxv. (1814), p. 351.

than 1 centim., and in places a bare 3 millim. in thickness. The hollow shell thus formed by decomposition is now entirely filled with quartz; and it is, indeed, questionable whether this relatively large cavity ever existed as a whole, it being perhaps the result of successive excavation and infiltration.

Seen in section, the rock is often traversed by rudely parallel cracks, some of which are posterior even to the infilling of the hollows of the altered spherulites. These cracks, filled with chalcedony, are remarkably wide wherever they traverse spherulitic matter, but become mere lines, for the most part, while passing through the matrix. Nor is this due to their being disguised by the secondary devitrification of the glass*, since the perlitic curves across which they occasionally run exhibit no such break as a wider fissure would have caused (Pl. IX. fig. 2). If, as is highly probable, the variation in width of these cracks has been determined by decomposition along their walls, their sudden expansion when they enter even the smaller spherulites is somewhat striking evidence of the greater stability of the matrix, and would lead us rightly to infer that the agents which excavate cavities in such rocks would produce a marked effect on the spherulites alone†.

The rocks of Saxony and the Wrekin area prepare us, then, for finding a mass composed of great nodules of quartz, each enclosed in a felsitic envelope, the products of alteration being thus out of all proportion to such primary structures as remain. As we have on a small scale spherulitic rocks in which the segregations outcrowd the matrix, so we have in the Silurian rhyolite of Digoed‡, near Penmachno, a mass some 15 feet thick, consisting almost entirely of white close-set nodules. These reach 7 or 8 centim. (3 inches) in diameter, and are almost all filled with quartz, the outermost layer of the spherulite being often alone preserved. A chloritic mineral is generally associated with the quartz (Pl. IX. fig. 3). Here and there the spherules suddenly diminish in size until their exceptional character is lost, while the matrix for a short interval assumes its normal predominance. Prof. Bonney§, to whose detailed investigation and description of North-Welsh nodular felsites students of this area must always turn, has referred to this rock in connexion with similar

* The product of this devitrification seems very largely to be felspar, twinning even being exhibited by many of the granules. Cf. Bonney, *Proc. Geol. Soc.* 1885, p. 94.

† It may be worth while to state the exact position of the quarry where the best specimens of the Lea Rock lavas seem always to have been obtained, since there are few excavations in the area. The road from Wellington joins the main Shrewsbury road at a point called "Hay Gate" on the 1-inch Ordnance Map. 1½ mile west of this is a toll-house, marked T. P., on the left. A large new red mansion lies a little further on upon the right, and beyond its lodge is a small cross road. The left hand or southern portion leads at once into the quarry. The name Lea Rock does not now seem to be recognized in the district.

‡ The farm of Digoed is found on the 1-inch Ordnance Map by following the high road south-south-east from Bettws-y-Coed till a branch goes off to Penmachno above the Conwy Falls. Digoed is set high on the east side of this branch, about 1 mile towards Penmachno, and the nodular rock rises in one or two bosses near the house.

§ Q. J. G. S. vol. xxxviii. p. 289.

examples from the neighbourhood of Bettws-y-Coed. It is interesting also to find that, before 1852, Delesse * examined at Jermyn Street certain nodular specimens from Llanberis and Digoed †, and classed them accurately with the "globular porphyries" and pyromerides of many foreign localities. The beautiful plates, drawn from polished surfaces of large and altered spherulites, that accompany his descriptions are monuments of faithful and minute work done when section-grinding was unknown.

In spite of their great size, the material of which the spherules of Digoed are constructed appears to have been very finely granular, and perhaps almost glassy. The relics of an often minutely curving flow-structure ‡, and the occurrence of small crystals of felspar scattered here and there, recall the features of the large spherulites of Zwickau. But alteration has sometimes revealed an original concentric structure, the secondary quartz occupying only the alternate zones. In one of these cases there is beautiful evidence of the pressure and strain § to which this rock has been subjected even since the deposition of the secondary material. A series of parallel bands of liquid-enclosures, containing moving bubbles, has been developed in a curved zone of granular quartz; these bands pass continuously from grain to grain, and are reproduced on opposite sides of the dull grey centre of the nodule, being thus clearly due to one common cause, which has left no mark upon the felsitic zones (Pl. IX. figs. 4 & 5).

Ample signs of attempts to crush and cleave the rock are seen, however, in the compression of the spherulites and sometimes in the irregularly fissile character of the matrix, where it can be said to be truly present. A cleavage has been also set up here and there in a black alteration-product which occurs within the spherules, and which, from its striking contrast to the hard, white, highly silicated rock, has seemed to me to merit peculiar notice.

Prof. Bonney, in the paper referred to ||, has mentioned a dull flinty-looking, but soft substance as occurring among the secondary minerals in felsites near Bettws-y-Coed and on the Conway Mountain. In the rock of the latter locality the dark patches formed by it are unusually abundant, and their relation to the zones of alteration in the nodules is often beautifully seen. One flattened spherulite, measuring 18 millimetres in its longest diameter, consists of at least thirty concentric coats, half of them being converted into the black material. The wavy structure seen in the latter, under the microscope, seems, from its general persistence of direction in the different zones, to be due to an attempt at cleavage which has been resisted by the less yielding portions of the rock (Pl. IX. fig. 6).

At Digoed this dark substance occurs on a coarser scale, so as to

* "Recherches sur les Roches globuleuses." *Mémoires de la Soc. géol. de France*, 2me série, tome iv. (1852), pp. 315 and 327.

† These specimens may be seen in the rock-room of the Geological Survey Museum, numbered 382, &c., in the 3rd edition of the Catalogue.

‡ Cf. Bonney, *Q. J. G. S.* vol. xxxviii. p. 294.

§ See Judd, *Q. J. G. S.* vol. xli. p. 376.

|| *Q. J. G. S.* vol. xxxviii. pp. 290, 293.

resemble fragments of included slate, still showing a relation to the curving zones of the great spherules. Its origin is, however, much better indicated by the Conway specimens.

With a specific gravity of 2·77*, a hardness of about 2·5, and all the appearance of a metamorphosed clay, it became interesting to compare this decomposition-product with the parent rock. I suspect that the composition of both varies somewhat in different specimens, especially in the matter of silica, which is so largely accumulated in the cavities of the rock. The portion of the white matrix and spheroidal matter chosen was, so far as could be seen, free from secondary quartz; but the high percentage of silica shown probably over-represents its original proportion in the lava-flow.

	I.	II.	III.
	White spherulitic rock of Digoed.	Black layer in large spherulite, Digoed.	Average of six analyses of pinite (the extreme per- centages made use of are placed in brackets).
SiO ₂	83·08	50·75	47·8 (44·7–54·6)
Al ₂ O ₃	10·25	28·34	29·9 (23·6–32·4)
Fe ₂ O ₃	trace	3·63	5·7 (0·9–10·2)
CaO	·26	2·57	·7 (0·0– 2·4)
MgO	·09	1·85	1·8 (0·0– 3·4)
K ₂ O	1·78	6·21	8·7 (6·5–11·2)
Na ₂ O	3·58	2·19	·5 (0·0– 1·1)
Loss on ignition ..	·74	4·37	5·1 (1·2– 7·8)
	99·78	99·91	100·2

I have placed in a third column the average of six analyses of pinite quoted by Rammelsberg †. It will be seen that the figures of the second column generally fall within the extremes made use of in calculating those of column III., the main difference being in the proportions of the alkalies.

The alteration-product of Digoed (and, by inference, the black material of the other ancient rhyolites) seems, then, to be related to the substances of the pinite group, and has probably as little claim as they to be regarded as a mineral species.

Under the higher powers of the microscope, moreover, it is seen to be composed of particles of various kinds. In one nodule from Digoed it has given rise to minute crystalline tufts and bunches of a uniform character ‡, interspersed with secondary quartz (Pl. IX. fig. 7); but a large section of the fragment used for the analysis consists mainly of transparent angular grains of a very pale blue-green

* The specific gravity of the white matrix rises as high as 2·67, but is variable through included quartz.

† Handbuch der Mineralchemie (2nd edit.), Band ii. p. 656.

‡ The substance discussed by Kalkowsky ("Untersuch. von Felsiten und Pechsteinen Sachsens," Tscherm. Mittheil. 1874, p. 46) may perhaps be of a similar nature.

tint, averaging .01 of a millimetre in diameter, together with a number of minute greenish prisms, which give vivid colours under crossed nicols. The latter are probably pyroxenic.

The results of the foregoing observations may be briefly summarized. Leaving out of count the matter added to an altered spherulitic rock by infiltration, such as the quartz which has, in lavas above the Conwy Falls, produced veins some six or seven inches wide, there is an evident tendency in such masses for the constituents to rearrange themselves, probably under the influence of heated waters as in the classical experiments of Daubrée*. The silica thus accumulates locally, and crystallizes out in nodular forms determined by the original spherulites, while materials of much more basic character than might have arisen from primary devitrification are developed in layers and patches, and prepare the rock for future physical change. It is rash to speculate on the final condition of such a rock, were it to come under the influence of mountain-building processes. Great "eyes" of quartz, however, resolvable under polarized light into granular aggregates, against which wisps of soft micaceous matter are pressed and bent, are features not unfamiliar in highly siliceous schistose rocks; and it seems probable that this differentiation and grouping of constituents would be closely imitated by further metamorphism of the lavas of Conway and Digoed.

The consideration of the changes undergone by our own coarsely spherulitic rocks leads one inevitably to the conclusion that the bulk, at least, of the "pyromerides"† and nodular porphyries of the continent were also originally glassy rocks. Prof. Bonney has practically expressed this view in dealing with the petrosiliceous structure in his Presidential Address for 1885‡. While it is of doubtful advantage to theorize respecting rocks with which one has no field-acquaintance, a few points of evidence may perhaps be mentioned here. Vogelsang§ states that in a dyke of "Kugelporphyr" at Ozani, Corsica, the spherulites are accumulated at the sides, an observation that may be paralleled on a minute scale in the glassy selvages even of basaltic rocks, as, for instance, in a vein near Tollymore, County Down. Delesse||, again, insists strongly on the connexion between an excessive proportion of silica and the development of "globular" or coarse spherulitic structure; and it need scarcely be pointed out that this same predominance of silica characterizes the most glassy of modern volcanic rocks. The following analysis by Delesse¶ of a spheroid from the pyromeride of Wuenheim, in the Vosges, may be compared with that of the rock

* Etudes synthétiques de géologie expérimentale, p. 159, &c.

† The name Pyroméride ("only in part fusible") is due to Haüy, and refers to the different behaviour of the constituents, quartz and "felspar." (Monteiro, Journal des Mines, tome xxxv. (1814), p. 359.)

‡ Proc. Geol. Soc. 1885, p. 95.

§ Neues Jahrb. für Min. &c. 1863, p. 102 (Niederrhein. Gesellsch. für Natur- und Heilkunde, Aug. 6, 1862).

|| Mémoires de la Soc. géol. de France, 2me série, tome iv. pp. 325, &c.

¶ Bull. de la Soc. géol. de France, 2me série, tome ix, p. 176.

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of Digoed. The silica-percentage has again, however, been probably raised by the removal of bases in solution. The specific gravity was 2.59.

Silica	88.09
Alumina	6.03
Oxide of Iron	0.58
Lime	0.28
Magnesia	1.65
Potash.....	} 2.53 (by difference)
Soda	
Loss on ignition	0.84
	100.00

De Lapparent*, again, has described a very striking pyromeride from Bouley Bay, Jersey, the spherules, which exhibit the characteristic infillings of chalcedony and quartz, being 25 centim. (10 inches) in diameter; but there seems to be little doubt, from an examination of specimens of several more modest varieties occurring in the same neighbourhood, that this rock also must be numbered among the ancient rhyolites†. Perlitic structure is, moreover, known to occur in the matrix of pyromerides, as at the Rauhfels of Wuenheim‡ and Gargalong near Fréjus§; and M. Michel Lévy||, while discriminating between the characters of acid rocks of different geological ages, points out, with Delesse, that there are considerable resemblances between many of the ancient "porphyries" and the rhyolites of Tertiary days. Whatever the issue in particular cases, the study of the slow processes of change to which igneous masses, in common with all other rocks, are subject, cannot fail to throw a vast amount of light on relationships hitherto obscured.

I gratefully record my indebtedness to Prof. Judd for kindly given help and much illustrative material; also to Mr. F. T. S. Houghton, F.G.S., for guidance at Conway and for several microscopic slides; and to Mr. J. F. Brooks, who worked with me in the Wrekin area, and who has checked some of the chemical results. Very many of the microscopic preparations referred to have been made in the Geological Laboratory of the Normal School of Science and Royal School of Mines, in which the work of analysis was also carried out.

* "Note sur les roches éruptives de l'île de Jersey." Bull. de la Soc. géol. de France, 3me série, tome xii. p. 287.

† Cf. T. Davies, "Old Rhyolites from Bouley Bay." Min. Mag. vol. iii. (1880), p. 118.

‡ Rosenbusch, 'Mikrosk. Physiographie' (1877), Band ii. p. 83. Delesse, Mém. de la Soc. géol. de France, 2me série, tome iv. p. 309.

§ Fouqué and Lévy, 'Min. micrographique,' plate xv. Lévy, "Roches anciennes acides." Bull. de la Soc. géol. de France, 3me série, tome iii. pp. 224, 235.

|| Loc. cit. p. 226.

EXPLANATION OF PLATE IX.

[Where the degree of enlargement of the objects drawn is expressed by a fraction, the numerator represents the magnifying-power of the objective with which they were viewed.]

- Fig. 1. Section of end of ramification of infilled cavity in large spherulite, pitchstone of Zwickau, showing its connexion with a crack. The lines of flow are cut through by the cavity, which contains chalcedony and calcite. A portion of the glassy matrix is seen below. $\times \frac{2}{3}$.
2. Section of coarsely spherulitic rhyolite, Wrockwardine (Wrekin) area, showing difference in width of cracks as they pass from the matrix into the spherulitic matter. The matrix is seen to be perlitic. $\times \frac{2}{3}$.
3. Large compressed spherulite, broken across, from rhyolite of Digoed, N. Wales. Extensive development of quartz and chlorite has taken place, and the residual spherulitic matter forms but a thin encircling wall. Natural size.
4. Section of two spherulites from rhyolite of Digoed, N. Wales, showing zones of granular secondary quartz, with lines of liquid enclosures passing continuously from grain to grain. $\times 2$.
5. Portion of the quartz of the above with lines of liquid-enclosures. $\times \frac{1}{2}$.
6. Section of compressed spherulite in rhyolite of Conway Mountain, showing alternate concentric layers composed of a black product of alteration. $\times 2$.
7. Section of black alteration-product in centre of spherulite, rhyolite of Conway Mountain, showing development of tufts of crystallites within it. $\times \frac{4}{3}$.

DISCUSSION

The PRESIDENT said that Mr. Cole had attacked a very puzzling problem and had given an explanation which appeared to be consistent with the whole of the phenomena.

Prof. BONNEY said that the subject was not only difficult to elucidate, but also difficult to discuss. He did not understand the formation of the supposed alteration-products from the white spherulitic rock. Again, he had noticed in one of the microscopic slides a crystal of iron-oxide close to the edge of a cavity. If the cavity was due to decomposition, why was not the crystal affected? Another difficulty was to conceive what agent could have produced the decomposition, what acid could have acted on one part of the rock, leaving other parts quite unaffected. He was rather inclined to attribute this kind of spheroidal structure to cracks forming around vesicles. The radial or true spherulitic structure was, in some cases, connected with, but subsequent in origin to, the formation of the cavity. He, however, congratulated Mr. Cole on the ingenuity of his views and on the lucid manner in which he had explained them to the Society.

Mr. RUTLEY said that he was unable to accept any view hitherto offered as to the origin of these bodies. He showed that there were several different kinds of spherulites, and gave examples. He instanced the concretions in the Magnesian Limestone of Durham as affording structure which sometimes apparently bore a certain

3.

nat. size.

analogy to that occurring in the large spheroidal bodies met with in the felsitic lavas of Skomer Island, and also in the forms which Mr. Cole had so well described.

Prof. MASKELYNE remarked that the solvent would more probably have been in the form of alkaline thermal infiltration than acid; but he did not understand how the black mineral was produced by simple decomposition of the white spherulitic rock. He doubted the data being sufficient to explain the action of alteration. For instance, what was the 4.37 per cent. loss by ignition? Was it all water or part of it carbonic acid? He suspected that while rocks like pitchstones differ in different districts, not merely in chemical, but in mechanical composition, the original formation of spherulitic structure was in most cases the result of contraction, owing to the minerals formed in the globules being denser and tending to shrink from the surrounding mass while solidifying.

Mr. COLE, in reply, said he had been greatly indebted to Prof. Bonney's papers. With reference to the alteration of the white spherulitic rock into the black product, the rock itself was not now in its original condition, and the additional alkalis of the product may have accumulated from the surrounding mass. Doubtless disintegration and infiltration had both taken place. He agreed that alkaline waters may have shared in the action. As to the hollowing of the spherulites, he referred to evidence put forward on the continent.